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Fuel Cycle Research and Development

Evaluation of Analytical Results Following Laboratory Scale Feasibility Study Voloxidation

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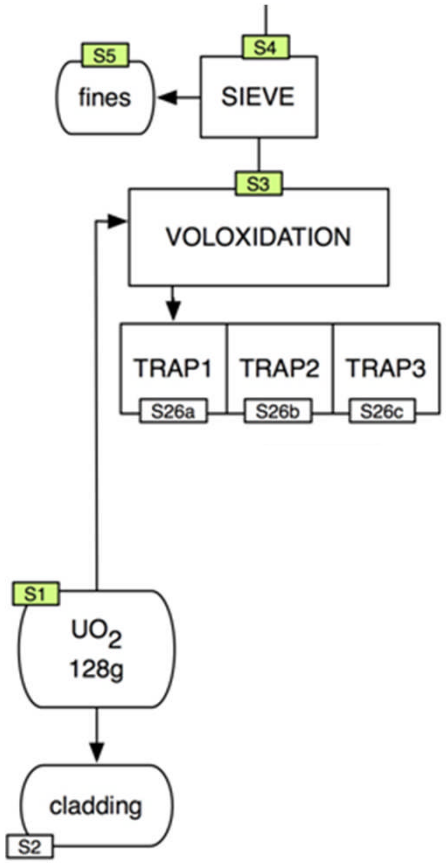
Outline

- **Laboratory-Scale Feasibility Study (LSFS) Sampling Plan**
  - *S1 is Pre-Test Fuel*
  - *S2 is Pre-Test Clad*
  - *S3 is Post-Test Fuel (Pre-Sieve)*
  - *S4 is Post-Sieve Coarse (>45 micron)*
  - *S5 is Post-Sieve Fines (<45 micron)*

- **Voloxidation**
  - Mass Balance/Sieving Results
  - Clad Analytical Chemistry Results
  - Fuel Analytical Chemistry Results
  - Filter Analytical Chemistry Results



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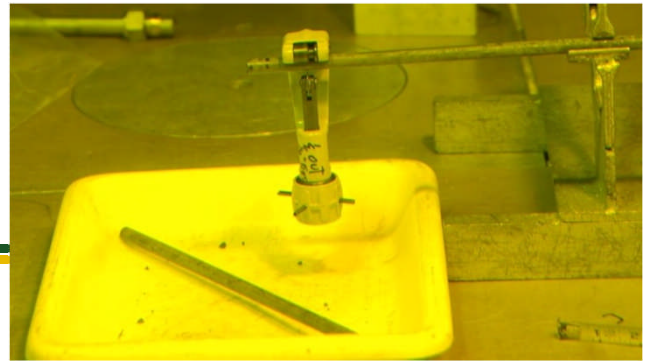
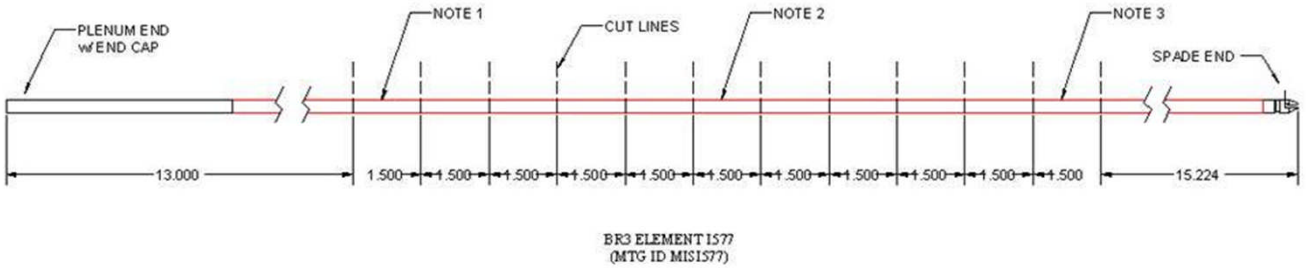
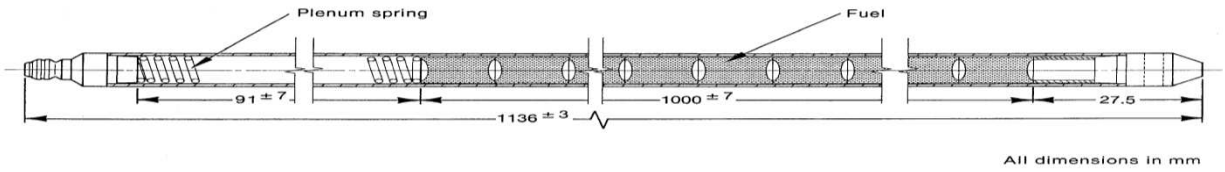
Voloxidation - Fuel Preparation



- **BR-3 Element I-577 (~41 GWd/t and 30 year decay)**
  - Eleven 3.81 cm (1.5”) segments sectioned from the center of the fuel region for a total of 41.91 cm of fuel
  - Segments crushed with impact mortar for decladding
  - 214 g of crushed fuel before sampling
  - 207 g of crushed fuel after sampling (S1)
  - Notes 1, 2, and 3 represent cladding samples (S2)



BR3 ELEMENT I577
(MTG ID MIS1577)



All dimensions in mm

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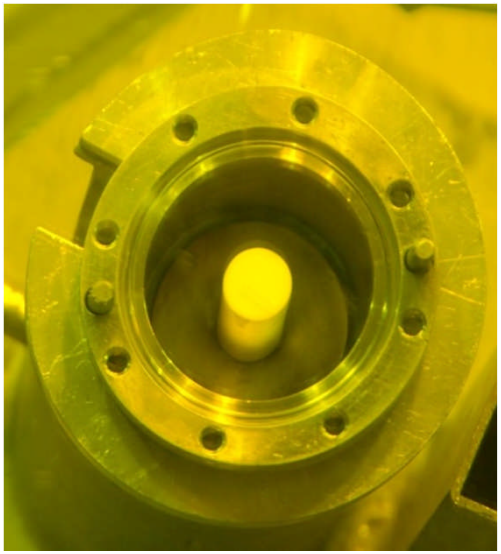
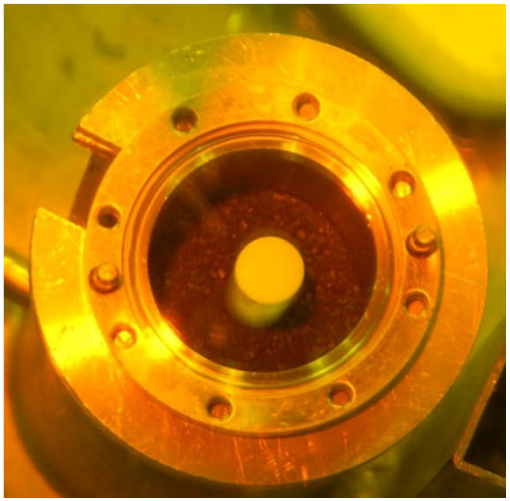
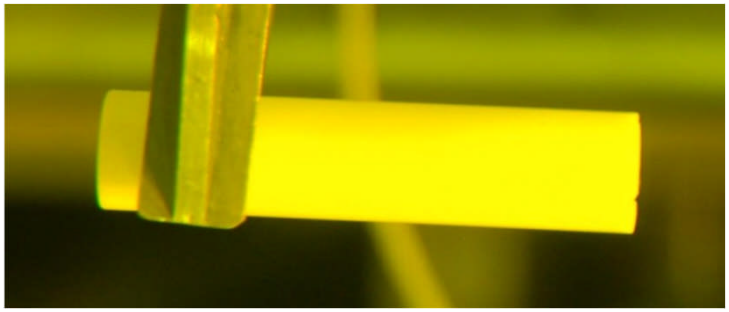
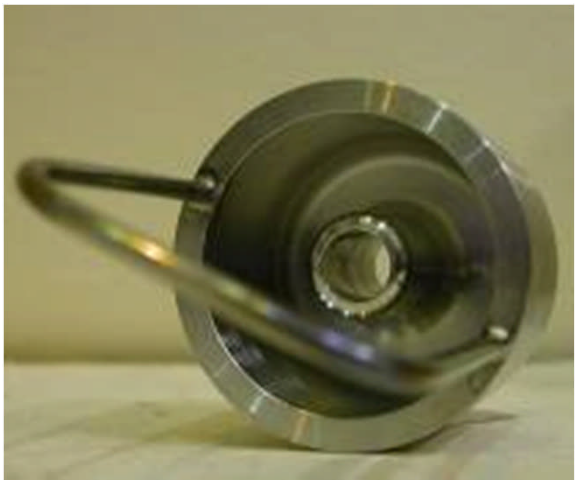
Voloxidation – Crucible Loading





- **Crushed fuel loaded into inner crucible**
  - Filled crucible to top
  - Bulk density of ~5.3 g/cc
- **Crushed fuel then loaded directly into outer crucible after slotted alumina shroud (1.73 cm OD x 7.62 cm H) to permit flow of air through fuel bed**





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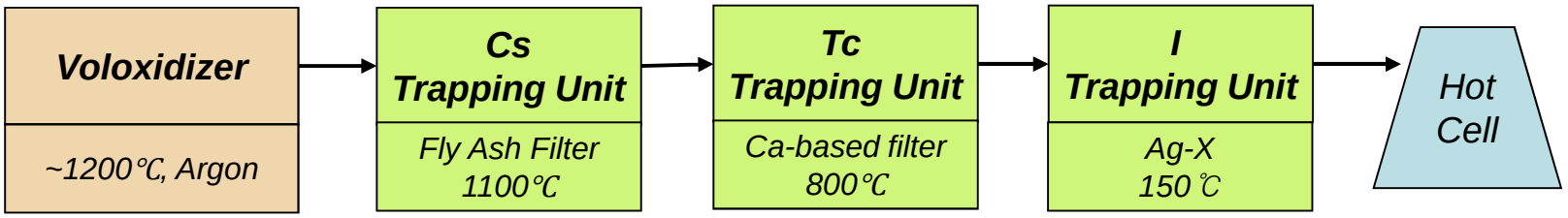
Voloxidation – Off-Gas Treatment System (OTS) Assembly

- **Filters Provided by KAERI**
  - Mesh type
    - *2.54 cm diameter by 1.8 cm height*
    - *AgX zeolite for iodine*
  - Disk type
    - *2.54 cm diameter by 1.3 cm height*
    - *Fly-ash based for cesium and technetium*

- **OTS Assembly**
  - Filters weighed
  - O and C-rings installed
  - Entire OTS leak checked
  - Sent to HFEF for transfer into hot cell



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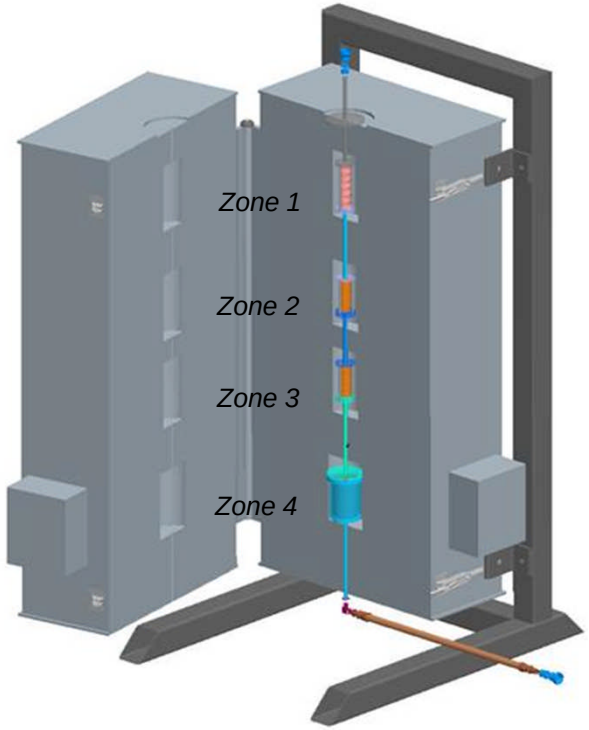
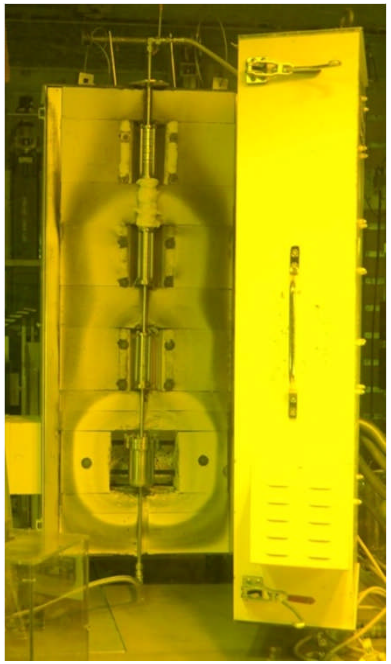
Voloxidation – Operating Conditions (1)



- **Zone 1**
  - Iodine collection with AgX filters
  - Preheat to 200°C for at least 12 hours prior to run to dehydrate zeolite
  - 150°C for entire run
- **Zone 2**
  - Technetium collection with CaO/fly-ash filters
  - 800°C for entire run
- **Zone 3**
  - Cesium collection with fly-ash filters
  - 1100°C for entire run
- **Zone 4**
  - Fuel oxidation with air at 1 scfh and 500°C for 3 hours
  - Disconnect air, connect Argon at 1 scfh, and heat to 1200°C for 5 hours to remove volatiles
  - Discontinue heat but continue heating Zones 1-3 until Zone 4 reaches 300°C



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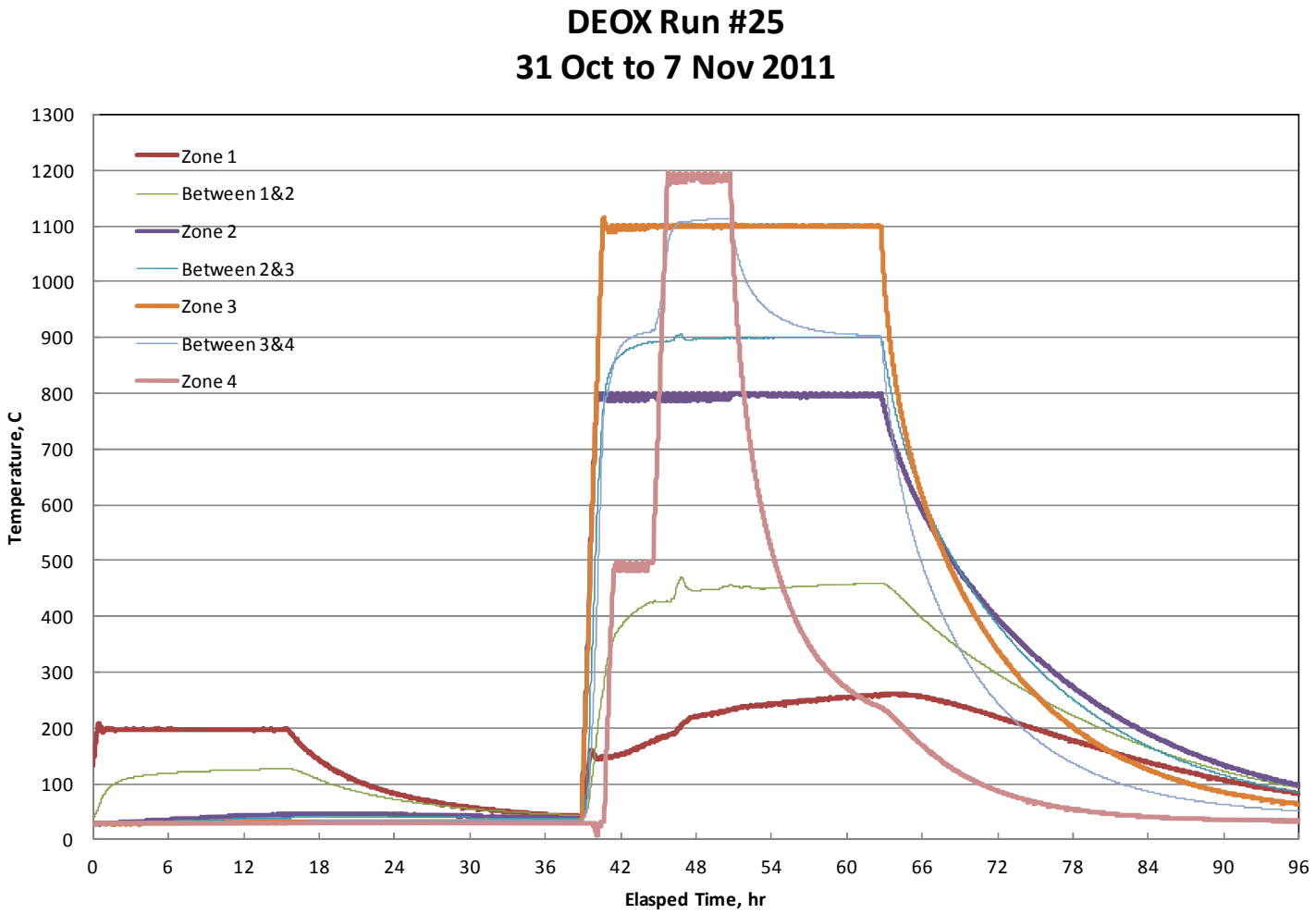
Voloxidation – Operating Conditions (2)

- ~15 Hour Dehydration of Zeolite Filters
- Zone 1 Maximum Temperature was 260°C
- Noticeable Temperature Spike in Tubing Between Zones 1 and 2



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Mass Balance/Sieving Results

Size Fraction	Quantity (g)	% Retained	Cumulative % Passing
>4 mm	3.4	1.8	100.0
2.8-4 mm	12.2	6.5	98.2
1.2-2.8 mm	42.1	22.4	91.7
0.6-1.2 mm	16.0	8.5	69.2
0.15-0.6 mm	18.3	9.8	60.7
.045-0.15 mm	53.0	28.3	51.0
<45 um	42.6	22.7	22.7

- **Sieving: 77% >45 micron**
- **Weight Measurements**
  - S1 weight after sampling = 207g
  - S3 weight before sampling (Pre-Sieve) = 203g
  - S3 weight after sampling (Pre-Sieve) = 195g
  - S4 weight before sampling (Post-Sieve) = 145g
  - S4 weight after sampling (Post-Sieve) = 132g
  - S5 weight before sampling (Post-Sieve) = 43g
  - S5 weight after sampling (Post-Sieve) = 30g
- **Mass Balance**
  - Voloxidation: Loss of 4 g (207-203)
  - Sieving: Loss of 7 g (195-145-43)

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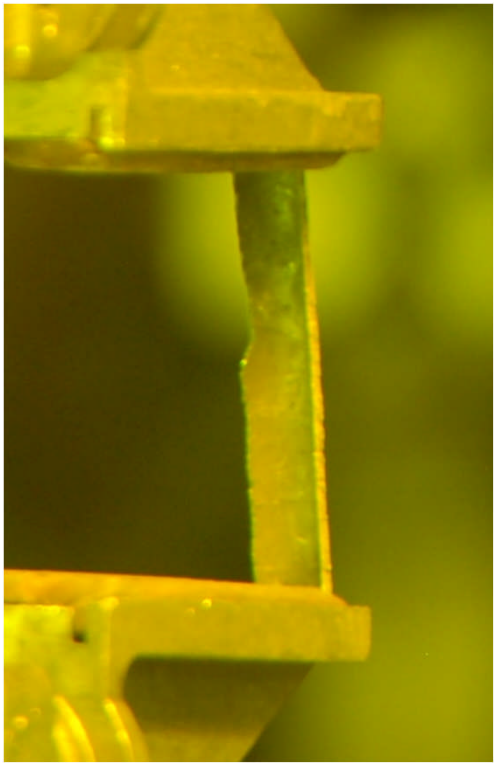
Cladding Results

- **Three cladding samples (S2) - 3.8 cm length**
- **Initial etch with HNO_3 followed by complete dissolution of clad with HNO_3/HCl/HF**
- **ICP-MS Performed: Isotopes of U, Np, Pu, Am, and Cm**
- **Results**
  - About 65% of initial etch is fuel (U, Np, Pu, Am, and Cm)
  - Calculate a 1-2 micron layer of clad/fuel etched
  - About 97% of fuel is in 1-2 micron layer
  - Measured amount of fuel retained in clad is <0.10 wt. % of total fuel for the 3.8 cm segment
  - Calculated alpha Pu (238, 239, 240) = ~10,000 nCi/g clad



Sample	Distance from Top (cm)	Initial Etch (mg)	Fuel in Etch (mg)	Fuel in Etch (%)	Fuel Total (mg)	Fuel in Etch (% of Total)
S2-1	33	12.8	7.5	58.5%	7.7	96.8%
S2-2	52	24.8	16.9	68.1%	17.3	97.5%
S2-3	71	20.3	13.7	67.4%	14.2	96.1%

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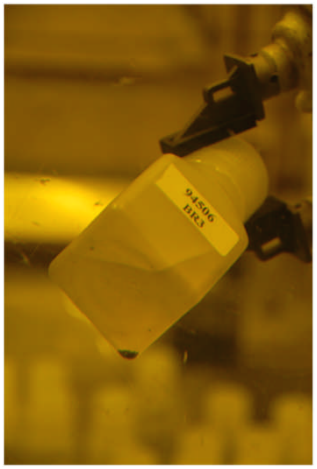
Fuel Results – Fission Products and Fe/Ni

Element	unit	S1-ave	S3-ave	S4-ave	S5-ave
Zr	ppm	5179	5211	5244	5124
Mo	ppm	3938	3153	3239	2795
Ru	ppm	2101	54	38	34
Rh	ppm	472	25	28	16
Pd	ppm	53	59	66	51
Tc	ppm	677	59	52	51
Nd	ppm	5416	5347	5352	5104
Ce	ppm	2885	2933	3043	2984
La	ppm	1634	1616	1633	1577
Pr	ppm	1398	1458	1442	1419
Sm	ppm	1044	1138	1206	1127
Eu	ppm	<130	<150	<130	<130
Sr	ppm	877	872	1007	849
Ba	ppm	2590	2503	2813	2030
Y	ppm	596	596	596	579
Te	ppm	553	295	347	288
Rb	ppm	478	270	290	273
Cs	ppm	2504	1019	1165	997
Fe	ppm	681	1642	355	<70
Ni	ppm	<420	5567	3820	561

- **Noble Metals (Mo, Ru, and Rh) affected by incomplete dissolution in Analytical Laboratory**



- **S1 vs. S3**
  - Tc removal of 91%
  - No significant change in Zr, Pd, Rare Earths (Nd, Ce, La, Pr, Sm, Eu, and Y), and Alkaline Earths (Ba and Sr)
  - Te removal of 47%
  - Rb removal of 44%
  - Cs removal of 59%
- **Less FPs in Fines Fraction (S5)**
- **Fe and Ni from Inconel corrosion**

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Fuel Results - Actinides

- Change in U Total caused by UO_2 ➔ U_3O_8 except S5 (Fines)
- Fines Fraction (S5) has more U and less TRUs (and less FPs) ➔ Burnup
- No significant change in TRUs from S1 to S3 (Am loss not observed)
- Pu Total/Cm244 for S4 = 3345

Actinide	unit	S1-ave	S3-ave	S4-ave	S5-ave
U Total	wt.%	80.65	76.81	77.23	80.66
U235	wt.% U	3.57	3.49	3.43	3.59
Np237	ppm	478	478	493	478
Pu Total	ppm	5879	5742	6021	5492
Pu239	ppm	4176	4072	4224	3932
Pu240	ppm	1420	1414	1498	1339
Pu241	ppm	81	51	71	43
Pu242	ppm	202	205	229	177
Am241	ppm	694	718	756	659
Am243	ppm	26	28	32	24
Cm244	ppm	1.4	1.6	1.8	1.3

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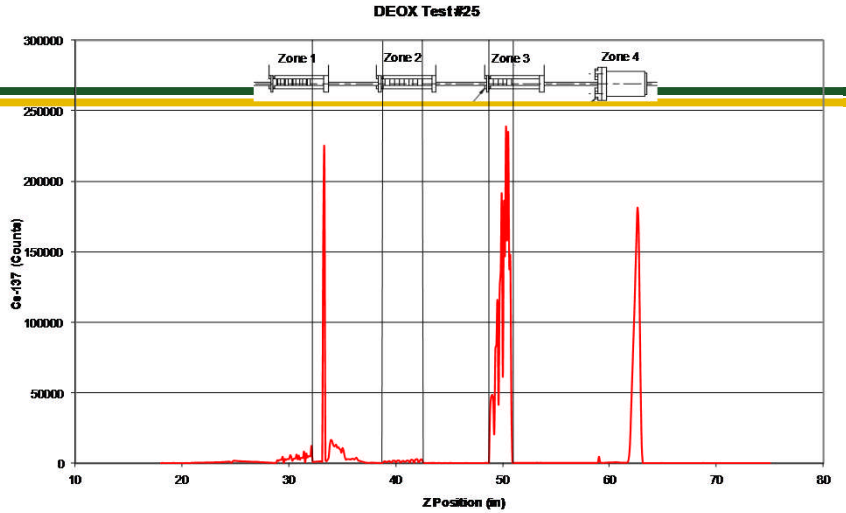
Filter Results – Zones 1, 2, and 3

- **Total Rb in Zones 2 and 3 is 32 mg (33%)**
  - Compare to 44% removal from fuel
- **Total Cs in Zones 1, 2, and 3 is 195 mg (38%)**
  - Compare to 59% removal from fuel
- **Total Tc in Zones 1, 2, and 3 is 3 mg (2%)**
  - Compare to 91% removal from fuel
- **Total I in Zone 1 is 32 mg (64%)**
- **Screen Sample from Zone 1**
  - 9.54 mg Cs
  - 4.90 mg Tc
  - 0.7 Tc:Cs (mol/mol)



Zone	Filter	I (mg)	Rb (mg)	Cs (mg)	Tc (mg)
3	1	NA	17.40	57.30	1.53
3	2	NA	7.81	52.70	0.29
3	3	NA	3.67	42.18	0.13
3	4	NA	2.06	26.58	0.07
3	5	NA	1.20	14.30	0.06
2	1	NA	0.07	0.52	0.28
2	2	NA	0.09	0.56	0.25
2	3	NA	0.06	0.31	0.24
1	1	31.93	NA	0.06	0.05
1	2	0.15	NA	0.06	0.03
1	3	0.05	NA	0.05	0.01
1	4	0.05	NA	0.04	0.01
1	5	0.03	NA	0.04	0.01

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Conclusions/Observations

- Although cladding retention of fuel was small (<0.10%), the TRU content would exceed 100 nCi/g criteria by 2 orders of magnitude
- Voloxidation is not necessary for homogenization of fuel, see S1 vs. S3
- Fines fraction post-voloxidation is a result of burnup (i.e. less burnup, more U content, thus better oxidation and pulverization of particles)
- Results of S3 and S4 very similar: ~75% of voloxidation product >45 um
- Slight reaction of voloxidation vessel (Inconel 600) was noted by presence of Ni in fuel samples due to extended time (5 hr) at 1200°C
- Incomplete removal of Cs, Rb, and Tc from fuel most likely due to lack of vacuum conditions at 1200°C during voloxidation
- Incomplete collection of Cs, Rb, and Tc on off-gas treatment system filters most likely due to deposit of material ($CsTcO_4$?) at the beginning of Zone 1
- Collection of iodine seems low although could be affected by material at beginning of Zone 1